

### Pulsewidth Dependence of Picosecond Laser-Induced Breakdown

E.W. Van Stryland, A.L. Smirl, and W.E. Williams

Department of Physics, North Texas State University,  
Denton, TX 76203, USA

M.J. Soileau

Michelson Laboratory, Physics Division, Naval Weapons Center,  
China Lake, CA 93555, USA

The laser-induced breakdown fields for fused  $\text{SiO}_2$ , single crystal NaCl and air were measured at  $1.06 \mu\text{m}$  for laser pulsewidths ranging from 40 psec to 31 nsec. These experiments represent the first such measurements over a range of  $10^3$  in pulsewidth while keeping other parameters such as specimen, laser frequency and focal volume constant. The laser-induced breakdown fields of fused  $\text{SiO}_2$  and single crystal NaCl were found to be only slightly dependent on pulsewidth, i.e.,  $E_B \sim t_p^{-x}$  where  $x \leq 1/8$ , whereas the air breakdown field varied as  $E_B \sim t_p^{-4}$ . The NaCl and  $\text{SiO}_2$  results are in sharp contrast to the predictions of various models [1,2] and previous published pulsewidth dependence data [2,3]. The work of BETTIS et al. [1] predicts that the breakdown threshold field will scale as  $t_p^{-4}$ . In contrast, work by SMITH et al. [3] indicated an approximately  $t_p^{-4}$  dependence of the breakdown field for NaCl using pulses in the picosecond region. The latter workers compared their 30 psec data with previous data from FRADIN et al. [4]. SMITH et al. [3] attempted to correct their data for self-focusing effects by using the technique proposed by ZVEREV and PASHKOV [5] but did not similarly reduce the data from FRADIN et al. [4], even though the latter authors exceeded the critical power for self-focusing by as much as an order of magnitude. In addition, the measurements performed by these two groups were taken with different focal volumes. Recent work has shown that in many cases laser-induced breakdown is dependent on focal volume and varies greatly among specimens of a given material [6,7]. Thus, pulsewidth dependence data is difficult to interpret unless all other parameters are held constant and the same specimen is used at all pulsewidths.

The laser source for the picosecond data presented in this work was a passively mode-locked Quantal, Nd:YAG system operating at  $1.06 \mu\text{m}$ . A single pulse from the mode-locked train was switched out and amplified to produce single pulses of measured Gaussian intensity profile and Gaussian temporal distribution. The temporal pulsewidth was variable between 30 and 200 psec by selecting various etalons as the output coupler. The width of each pulse was determined by monitoring the ratio, R, of the square of the energy in the fundamental ( $1.06 \mu\text{m}$ ) to the energy in the second harmonic produced in a  $\text{LiIO}_3$  crystal. This ratio is directly proportional to the pulsewidth and was calibrated by measuring the pulsewidth using second harmonic autocorrelation scans and accepting only a narrow range in ratios, R. Damage data was then selected according to the temporal width of individual pulses. The breakdown threshold was taken to be that intensity at a given pulsewidth which produced damage 50% of the time. Each site was irradiated only once. The energy on target was varied by changing the angle between a calibrated pair of Glan polarizers. The nanosecond data was taken on the same samples using a Q-switched Nd:glass laser. The same focusing lenses were used, and the energy monitor was directly calibrated with the one used for the picosecond measurements.

AIR			SiO <sub>2</sub>			NaCl		
$\omega_0$ ( $\mu\text{m}$ )	$t_p$ (psec)	$E_B$ (MV/cm)	$\omega_0$ ( $\mu\text{m}$ )	$t_p$ (psec)	$E_B$ (MV/cm)	$\omega_0$ ( $\mu\text{m}$ )	$t_p$ (psec)	$E_B$ (MV/cm)
19.3	31,000		19.3	31,000		19.3	31,000	2.16 $\pm$ 0.22
	191 $\pm$ 26	36.8 $\pm$ 2.3		133 $\pm$ 13	8.4 $\pm$ 0.4		134 $\pm$ 13	2.68 $\pm$ 0.22
	104 $\pm$ 16	40.8 $\pm$ 3.0		76 $\pm$ 7	9.2 $\pm$ 0.6		92 $\pm$ 8	2.86 $\pm$ 0.13
	40 $\pm$ 3	55.8 $\pm$ 6.2		40 $\pm$ 2	8.4 $\pm$ 0.3		42 $\pm$ 3	2.74 $\pm$ 0.17
10.3	31,000	14.2 $\pm$ 1.4	10.3	31,000	8.38 $\pm$ 0.8	10.3	31,000	2.67 $\pm$ 0.26
	183 $\pm$ 17	42.2 $\pm$ 1.8		176 $\pm$ 12	12.3 $\pm$ 0.6		176 $\pm$ 26	3.29 $\pm$ 0.39
	117 $\pm$ 17	61.6 $\pm$ 3.4		78 $\pm$ 6	12.6 $\pm$ 1.0		117 $\pm$ 8	3.84 $\pm$ 0.30
	47 $\pm$ 6	67.4 $\pm$ 3.3		47 $\pm$ 6	12.3 $\pm$ 1.0		47 $\pm$ 6	4.20 $\pm$ 0.30
6.1	31,000	20.8 $\pm$ 2.0	6.1	31,000	11.4 $\pm$ 1.1	6.1	31,000	2.83 $\pm$ 0.29
	128 $\pm$ 8	84.3 $\pm$ 2.1		176 $\pm$ 20	16.0 $\pm$ 1.2		167 $\pm$ 16	4.91 $\pm$ 0.30
	106 $\pm$ 10	86.0 $\pm$ 3.4		100 $\pm$ 16	16.8 $\pm$ 1.1		100 $\pm$ 10	6.80 $\pm$ 0.40
	43 $\pm$ 2	88.1 $\pm$ 2.2		47 $\pm$ 6	16.7 $\pm$ 1.0		48 $\pm$ 3	8.08 $\pm$ 0.37
5.0	31,000	28.8 $\pm$ 3.0	5.0	31,000	12.6 $\pm$ 1.3	5.0	31,000	
	134 $\pm$ 8	93.1 $\pm$ 2.8		134 $\pm$ 8	26.9 $\pm$ 1.6		128 $\pm$ 16	8.10 $\pm$ 0.8
	100 $\pm$ 8	96.6 $\pm$ 3.4		100 $\pm$ 8	28.8 $\pm$ 1.8		104 $\pm$ 7	8.30 $\pm$ 0.4
	44 $\pm$ 2	136 $\pm$ 4.0		43 $\pm$ 3	30.1 $\pm$ 1.2		43 $\pm$ 3	11.60 $\pm$ 1.4

Fig. 1. A tabulation of the RMS breakdown electric fields,  $E_B$ , of SiO<sub>2</sub>, NaCl and air for focused spot sizes ( $1/e^2$  half-width in intensity) of 5.0, 6.1, 10.3, and 19.3  $\mu\text{m}$  and for various optical pulsewidths (FWHM) in psec

The results of our measurements over a range of  $10^3$  in pulsewidth for focal spot radii from 5.0 to 19.3  $\mu\text{m}$  are summarized in Fig. 1. Self-focusing effects were neglected in calculating the breakdown fields. Some workers claim that observed dependences of breakdown fields on focal spot radii are due to self-focusing [3,5], and they scale their results in accordance with the technique suggested by ZVEREV and PASHKOV [5]. ZVEREV and PASHKOV [5] predict that a plot of  $P_B^{-1}$  ( $P_B$  is the power at which breakdown occurs) versus  $\omega_0^{-2}$  yields a straight line given by

$$P_B^{-1} = 2(I_B \pi \omega_0^2)^{-1} + P_{CR}^{-1} \quad (1)$$

where  $P_{CR}$  is the critical power for self-focusing and  $I_B$  is the breakdown intensity. The basic assumption of this procedure is that  $I_B$  is the intrinsic breakdown intensity and is independent of the focal spot radius. It is clearly seen from Fig. 2 that this scaling technique cannot be used for our experiments. All of the data for the different pulsewidths follows a similar pattern and cannot be fit to a straight line. Deviations from the straight line fit for large spot sizes have been noted previously [3]. Such data have been disregarded by arguing that for large focal radii and powers near  $P_{CR}$ , the constant shape solution to the nonlinear wave equation (on which the ZVEREV and PASHKOV [5] procedure is based) is no longer valid. However, the argument cannot explain the small focal radii data shown in Fig. 2.

It has been clearly established that with the possible exception of a small number of specimens tested by MAIENKOV [6], the laser-induced breakdown fields are not intrinsic, and vary greatly even for specimens of a given material from the same supplier [7,8,9]. This violation of the basic assumption that the damage is intrinsic casts doubt on previously published data where the ZVEREV and PASHKOV [5] scaling was used to interpret breakdown thresholds.

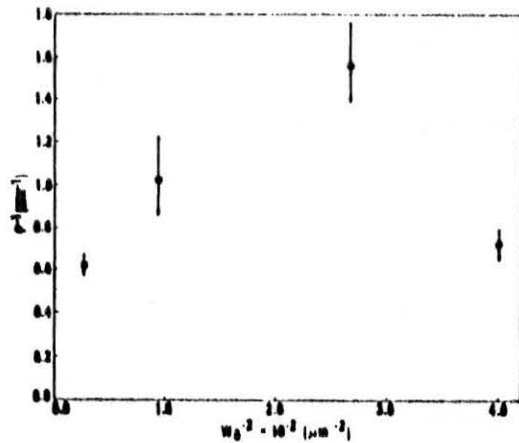


Fig. 2 A representative plot of the inverse of the power,  $P_B$ , needed to induce breakdown versus  $w_0^{-2}$ , where  $w_0$  is the  $1/e^2$  radius in intensity at the laser focus calculated using linear Gaussian optics. Here data for SiO<sub>2</sub> using 44 psec (FIHM) pulses is presented

We conclude that the role of self-focusing in these experiments cannot be determined, and therefore, the data presented in Fig.1 do not include any self-focusing corrections (even though  $P_B$  exceeds many estimates of  $P_{CH}$ ). This apparent discrepancy with self-focusing theory can be explained by including the effect of plasma defocusing. It has been shown that the negative nonlinear index of refraction,  $n_2$ , due to free electrons produced by the intense optical fields counters the positive  $n_2$  resulting from bound electronic effects [7,8].

The pulsewidth dependence as shown in Fig.3 is important for understanding the basic mechanisms of laser-induced failure. Only part of the data of Fig.1 is shown in Fig.3 but that shown is typical of all the data. Air breakdown was observed to scale as  $t_p^{-4}$  as predicted by the simple avalanche theory; however, bulk damage in both SiO<sub>2</sub> and NaCl show a much slower dependence on  $t_p$  than predictions of various models and previous [1,2] pulsewidth dependence data [2,3].

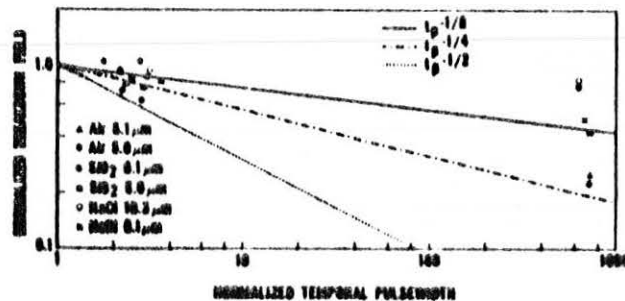


Fig.3 The normalized RMS breakdown field versus normalized temporal pulsewidth at 1.06  $\mu\text{m}$ . The breakdown field was normalized by dividing the field by the breakdown field at the shortest pulsewidth employed for that material and spot size. The pulsewidth was then scaled by the shortest pulsewidth. For example, the breakdown field for SiO<sub>2</sub> at a spot size of 6.0  $\mu\text{m}$  was determined by normalizing the breakdown field by 30.1 MW/cm and the pulsewidth by 43 psec (see Fig.1). This normalization assures that the intercept goes through 1.0

For example, the fit of data from [3] and [4] to predictions based on d.c. breakdown theory has been interpreted as evidence for the avalanche breakdown model. The  $t_p^{-4}$  dependence of the breakdown field determined in [3] was used to scale 30 psec data at 1.06  $\mu\text{m}$  to 21 psec so that frequency dependence between 1.06 and 0.53  $\mu\text{m}$  could be examined [10]. As a result of this scaling, the breakdown fields at 0.53  $\mu\text{m}$  are found to be greater than those at 1.06  $\mu\text{m}$  further supporting the avalanche model. However, if the scaling results for NaCl and SiO<sub>2</sub> shown in Fig.3 are used instead, the breakdown field at 0.53  $\mu\text{m}$  would be lower than at 1.06  $\mu\text{m}$  and would indicate that avalanche breakdown was not the damage mechanism.

There are laser-induced breakdown theories which predict very weak pulsewidth dependence similar to the bulk damage results presented here. For example, SPARKS [11] predicts a  $t_p^{-1/6}$  scaling of the laser-induced breakdown fields. BRAUNLICH, et al. [12] have pointed out that multiphoton processes should become increasingly more important as laser pulsewidth decreases. They predict that in the absence of an avalanche,  $t_p^{-1} \propto (E_B)^{n+1}$  where  $n$  is the order of the multiphoton process. The band gap of SiO<sub>2</sub> and NaCl is approximately 7.8 eV. Direct excitation of electrons across the gap would be a 7 photon process for those materials at 1.06  $\mu\text{m}$  for which case the pulsewidth dependence would be  $E_B \propto t_p^{-1/8}$ . Although it is unlikely that direct excitation of electrons across the gap is the mechanism for laser induced failure of NaCl and SiO<sub>2</sub>, this and previous [7,8] pulsewidth dependence studies indicate that multiphoton processes may play a more important role in laser-induced breakdown than was previously thought. The data of Fig.1 are qualitatively consistent with the multiphoton-initiated avalanche breakdown model presented in Refs. 7 and 8 [13].

This work was performed as part of a joint NTSU/NIIC research program sponsored by the Office of Naval Research (ONR) and the North Texas State Faculty Research Fund. A. L. Smirl also wishes to thank the ONR for support.

- 1 J. R. Bettis, R. A. House, and A. H. Guenther, NBS Special Publication 462, p. 338, 1976.
- 2 W. Lee Smith, Optical Engineering 17, 409 (1978).
- 3 W. Lee Smith, J. H. Bechtel, and N. Bloembergen, Phys. Rev. B 12, 796 (1975).
- 4 D. H. Fradin, N. Bloembergen, and J. P. Letellier, Appl. Phys. Lett. 22, 635 (1965).
- 5 G. M. Zverev and V. A. Pashkov, Soviet, Phys. JETP 30, 616 (1960).
- 6 A. A. Manenkov in NBS Special Publication 509, p. 445, 1977.
- 7 M. J. Soilleau, Ph.D. Thesis, University of Southern California, 1979.
- 8 M. J. Soilleau, M. Bass, and P. H. Klein, "Frequency and Focal Volume Dependence of Laser-Induced Breakdown in Wide Band Gap Insulators," in the NBS proceedings of the 1979 symposium on Laser Induced Damage to Optical Materials.
- 9 D. Olness, Appl. Phys. Lett. 8, 283, 1966.
- 10 W. Lee Smith, J. H. Bechtel, and N. Bloembergen, Phys. Rev. B 15, 4039 (1977).
- 11 H. S. Sparks, NBS Special Publication 435, p. 331, 1975.
- 12 P. Braunlich, A. Schmid, and P. Kelly, Appl. Phys. Lett. 26, 150 (1975).
- 13 E. W. Van Stryland, M. J. Soilleau, Arthur L. Smirl, William E. Williams, to be published.